A study of cloud processing of organic aerosols using models and CHAPS data

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Motivation

- Current aerosol model **significantly underpredict secondary organic aerosol (SOA) mass**
- Recent findings (model, lab, field) suggest that **chemical processes in cloud droplets** might lead to organic mass that remains in the particle upon cloud evaporation
- The efficiency of this SOA formation pathway **has not been quantified yet**

1. Mechanism development

- Lab experiments: aqueous phase reactions of organic species (carbonyls, acids)

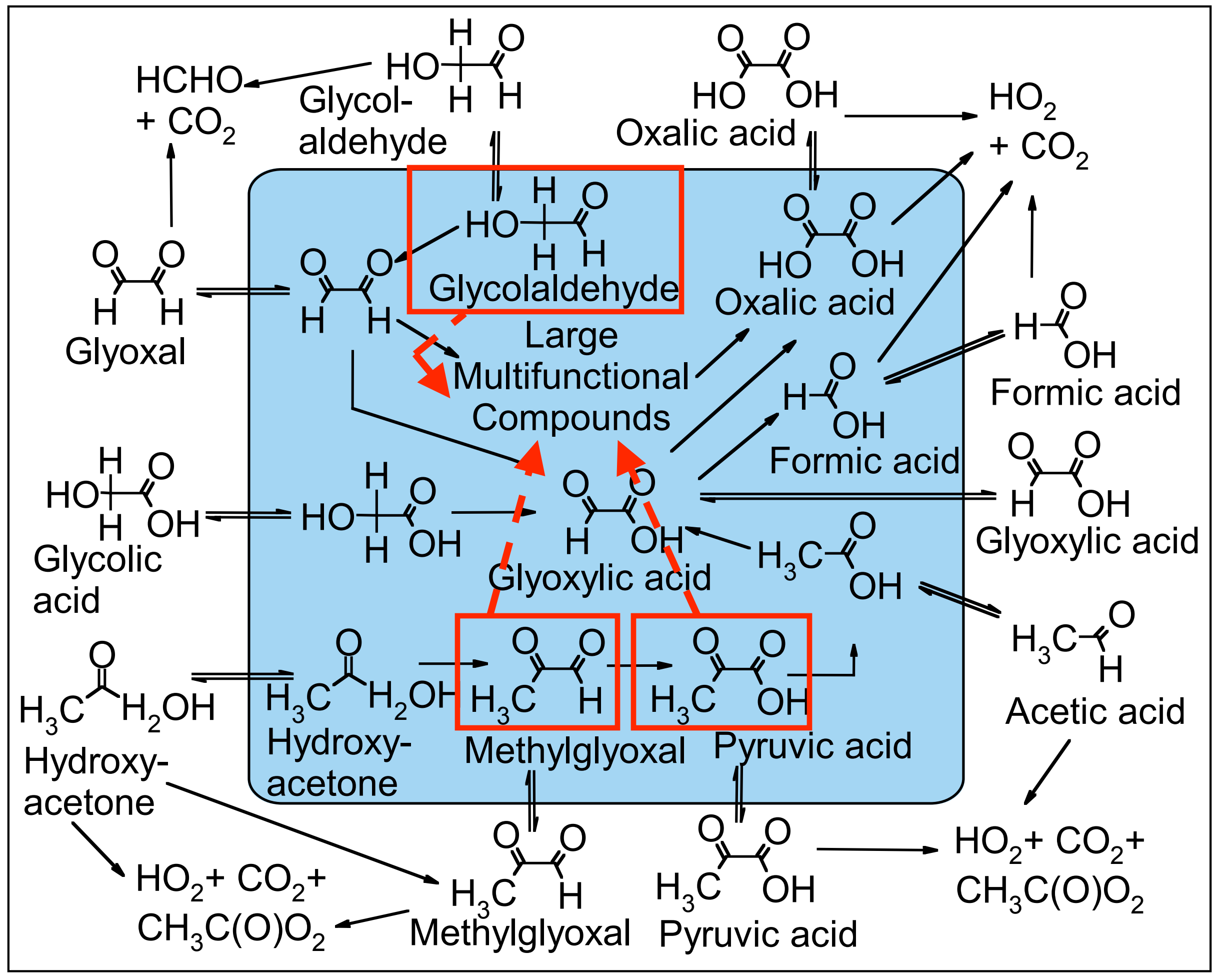
  Collaboration with B. Turpin (Rutgers University)

- Derivation of chemical mechanism (rate constants, species) by fitting of measured concentration profiles *Carlton et al., 2007, Altieri et al., 2008*

Extension of the current mechanism:

Detailed reaction scheme for
- methylglyoxal
- pyruvic acid
- glycolaldehyde

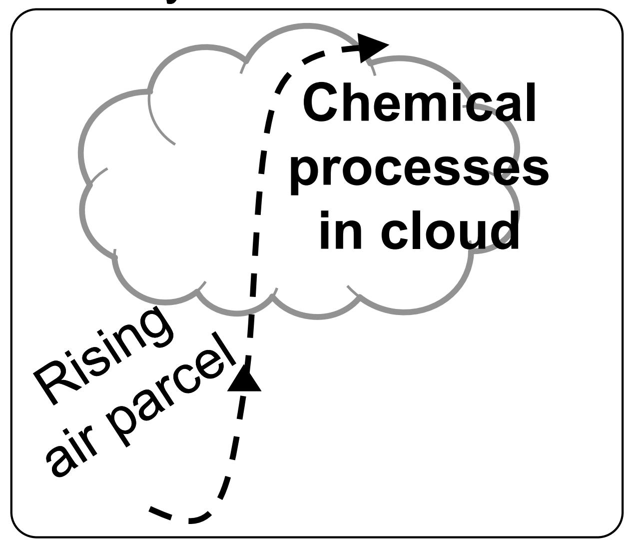
Interactions of organics with other compounds (e.g., sulfate)



2. Model application: CHAPS data

- **Cloud parcel model** with organic and inorganic multiphase (gas, aqueous) chemistry *Feingold et al., 1998, 2000; Ervens et al., 2004, 2008*

- Trajectories of suitable CHAPS cases will be derived by LES model (PNNL): Drivers for parcel model



Model initialization:

- Meteorological data
- Gas phase VOC
- Aerosol size distribution and composition

Closure studies:

Comparison of observed and predicted properties of initial vs. cloud-processed aerosol

- Sulfate mass, (speciated) organic mass in cloud residuals
- Optical properties
- CCN properties

mass (size) and composition modification

Collaboration with J. Ogren, NOAA

3. Parameterization of SOA_{drop} formation

- **Parameterization of SOA_{drop} yields**, in a similar way as it has been done for SOA from isoprene in stratocumulus clouds

$$Y(SOA_{drop}) = \frac{\Delta\,(\text{mass } SOA_{drop})}{\Delta(\text{mass precursor VOC})} \quad = \text{f(cloud parameters [LWC, } \tau\text{], } NO_x)$$

Only small dependence on pH, air/water interface, partitioning ratio

Ervens et al., Geophys. Res. Lett., 2008

- These parameterizations will be **implemented into a module that treats aerosol transformation** in fair weather cumuli for application in regional and global climate models (PNNL, M. Ovtchinnikov, R. Easter)

- **Refinements** of model input/parameterizations
  - *chemical* processes: e.g.,less dilute particles, SOA precursors, new lab results, ...
  - *meteorological* parameters: Is in-cloud aerosol connected with below/above cloud aerosol? (⇨ transport, mixing)